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Numerical Simulation of CALIBAN Reactivity Perturbation Experiments Using Neptunium-237 Samples

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ABSTRACT

In order to contribute to the validation of nuclear data used in critical mass computation, reactivity perturbation experiments using ^{237}Np samples have been performed at CEA-Valduc using the fast pulsed reactor CALIBAN operated in continuous mode. In this paper we report these experiments together with the numerical calculations.
The calculations were carried out using PANDA, a S_N code developed at CEA-Bruyères-le-Châtel for classic criticality and stochastic neutronics applications and with MCNP4C which is a commonly used Monte Carlo code.
A good agreement was found between experimental and deterministic results.

Key Words: fast pulsed reactor, perturbation, reactivity, neptunium, neutron transport codes

1 Introduction

Limited experimental data concerning the ^{237}Np critical mass can be found [2]. There are no direct evaluation using near-critical assembly. Nevertheless, reactivity perturbations measured in replacement experiments are sensitive to the sample nuclear data [1] *and* [3]. These experiments can contribute to validate nuclear data used in critical mass computation.

In this paper we present such an experiment using CEA-Valduc CALIBAN [4] fast reactor operated in continuous mode.

In a first part we present the CALIBAN critical assembly and the measurement technique used to determine the excess of reactivity due to the ^{237}Np samples and we report the experimental results. The following sections are concerned by the numerical simulations of the experiments.

2 Description of the Experiments

2.1 General

The experiments took place in the CEA Valduc center facility during the year 1993. They are close to the experiments described in the interesting paper of René Sanchez from LANL [1]. Small gram sized samples of ^{237}Np have been placed in the center of the cavity of a fast pulsed reactor operated in continuous mode. The characteristics of the CALIBAN reactor are close of those of SPRII (Sandia Pulsed Reactor II).

2.2 CALIBAN fast reactor

CALIBAN [4] is an unreflected fast HEU core reactor with a central cavity used for sample irradiation, it may be seen as a cylindrical reactor. It diverged for the first time in the beginning of the seventies and when used without reflector, its critical Uranium charge is close to one hundred kilograms.

The core of the reactor consists of circular discs and four rods. It is made of a 90 % Uranium (with an enrichment of 93.5 %) and 10 % Molybdenum alloy. These values are given in weight.

One of the rods serves for the rapid injection of the reactivity and initiation of the pulse, this rod is not used for the experiments described in this paper. The three other rods are disposed in cylindrical vertical channels parallel with the axis of the reactor.

CALIBAN reactor can be operated in continuous mode at delayed critical with a long asymptotic period, and so the measurement of the reactivity perturbations can be accurately measured and may be used as benchmarks.

2.3 Measurement technique

The reactivity worth of the samples has been measured in the following way :

The reactor is operated above delayed critical by inserting two of the three control rods to their full-in position. The third one is used to measure the reactivity worth of the sample. The measured asymptotic period is higher than thirty thousands seconds for each experiment, so the inhour equation for a HEU reactor gives a k_{eff} lower than 1.000003 but supercritical.

- Step 1 : Before measuring the reactivity worth of any sample, the third control rod is positioned to obtain an asymptotic period higher than thirty thousands seconds with no sample in the cavity.
- Step 2 : The iron can is then put in the cavity and the third control rod is positioned to have an asymptotic period higher than thirty thousands seconds. This step gives the reactivity worth of the iron can.
- Step 3 : The final step is done with the complete sample, that is gram-sized ^{237}Np (16.32 g, 32.66 g, 48.98 g) surrounded by the iron can. The reactivity worth is obtained with the help of the third control rod position corresponding with a long asymptotic period (higher than thirty thousands seconds).

2.4 Experimental results

The experimental results are given in table 1. The experimental uncertainties (Temperature, time period, ...) are taken into account. The specific worth is given in $10^{-5}/gram$ by :

$$w_{exp} = \frac{\rho_{sample} - \rho_{coating}}{M(^{237}Np)}$$

$$\rho = \frac{k_{eff} - 1}{k_{eff}}$$

TAB. 1 – **Neptunium-237 samples experimental specific worth.**

Sample Mass (g)	$\Delta\rho$ (10^{-5})	w $(10^{-5}/g)$
16.32	25.93 ± 0.1	1.589 ± 0.006
32.66	53.30 ± 0.1	1.632 ± 0.003
48.98	80.77 ± 0.1	1.649 ± 0.002

3 Computer Model

In order to perform calculations using numerical codes, 2D computer models of CALIBAN critical assembly with and without ^{237}Np sample and coating were designed. These models are axisymmetric with respect to Z axis and present a reflective symmetry at Z=0. Dimensions and material data concerning CALIBAN are given in table 3. An image of the different components of the assembly considered by the computer model is presented in figure 1. Three coated samples were measured. These samples and the corresponding empty coatings models characteristics are given in table 2.

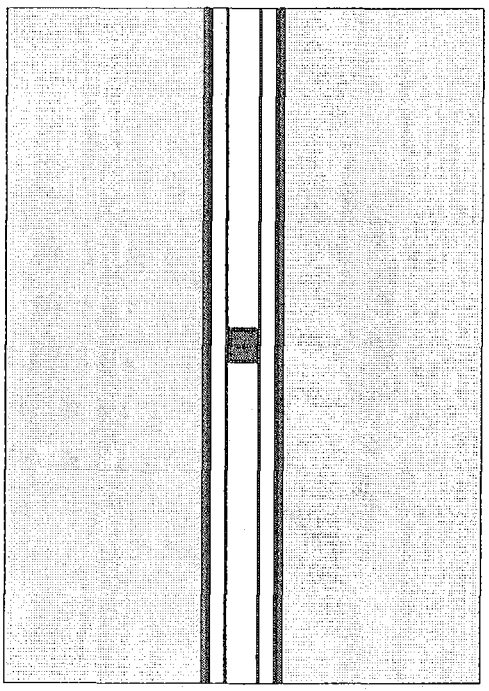

FIG. 1 – **CALIBAN with sample computer model: 2D-axisymetric and Z=0 reflective boundary condition. (HEU Core, Iron, Neptunium, void)**

TAB. 2 – **Samples and coatings models characteristics.**

	Uncoated	coatings
Isotopes ($at\%$)	^{237}Np (100%)	^{56}Fe (100%)
Density (g/cm^3)	20.45	7.90
R, H (cm)	R = 0.48 H = 1.10 2.20, 3.30	Thickness = 0.08
mass (g)	16.28 32.56 48.85	3.52 5.79 8.06

TAB. 3 – **CALIBAN model dimensions and material properties.**

	Core	Core internal cover	Sample holder guide	Sample holder
Isotopes ($at\%$)	^{235}U (73.46%) ^{238}U (5.13%) Mo (21.41%)	^{56}Fe (100%)	^{56}Fe (100%)	^{56}Fe (100%)
Density (g/cm^3)	17.04	7.90	7.90	7.90
Internal Radius (cm)	1.50	1.30	1.20	0.60
External Radius (cm)	8.75	1.50	1.25	0.65
Height (cm)	25.066	25.066	25.066	25.066

TAB. 4 – **16-Groups Energy Structure.**

Group number	Lower Energy (MeV)	Upper Energy (MeV)	Group number	Lower Energy (MeV)	Upper Energy (MeV)
1	0.001	0.010	9	3.000	5.000
2	0.010	0.030	10	5.000	6.000
3	0.030	0.100	11	6.000	8.000
4	0.100	0.200	12	8.000	10.000
5	0.200	0.500	13	10.000	11.000
6	0.500	1.000	14	11.000	12.000
7	1.000	2.000	15	12.000	13.000
8	2.000	3.000	16	13.000	15.000

4 Numerical Simulations

We have used ENDF BVI, JEF2 and JEF3 cross sections. We will show results with ENDF BVI data, and we give some words about the use of the other cross sections in the conclusion of this paper.

4.1 PANDA code

Deterministic simulation was performed using the PANDA [6] code. This code was developed at CEA-Bruyères-le-Châtel for criticality and stochastic neutronics applications. It is based on the classic S_N method. The transport equation is discretized using a finite volume method and a centered diamond scheme is used for angular and spatial variables. The input geometries can be described in one dimensional (plane, spherical and cylindrical) or two dimensional (plane and cylindrical) coordinates. The angular discretization uses S_N quadrature. The anisotropic scattering is handled by Legendre expansion for neutron flux and cross sections. The energy variable is discretized using the multigroup method. Fixed point iterative methods are used for flux and eigenvalue computations.

Regarding numerical calculation of the reactivity perturbation using deterministic code two methods have been considered. The first one uses the difference of two very precise k_{eff} calculations. The second method uses first order perturbation. This method assumes a linear behavior of the reactivity excess with sample mass.

4.1.1 Convergence parameter

The k_{eff} calculation is performed using an optimized eigenvalue search algorithm. This iterative algorithm proceeds by eigenvalue extrapolation in an automatically adjusted interval. This method is more efficient with PANDA than the classic "power iteration" method. The algorithm is converged for a given value of the ϵ parameter when both following unequalities using neutron flux norm for iteration number n and $n-1$ are verified.

$$\left|\frac{||\Phi^{(n)}||}{||\Phi^{(n-1)}||}-1\right| \leq \epsilon \quad \left|||\Phi^{(n)}||-1\right| \leq \epsilon$$

The flux norm is given by:

$$||\Phi|| = \int_V d^3r \int_0^\infty dE\ \Phi(\vec{r},E)$$

The convergence parameter was set to $\epsilon = 10^{-7}$ for the difference method and to $\epsilon = 10^{-5}$ for the perturbation method.

4.1.2 First order perturbation method

Using the first order perturbation theory the reactivity perturbation is given by [5]:

$$\delta\rho = \frac{(\varphi^+, [\delta F - \delta M]\varphi)}{(\varphi^+, F\varphi)}$$

where φ and φ^+ are the direct and adjoint unperturbed flux, M and F are the transport and fission operators and δM and δF are the perturbation operators.

4.1.3 Angular and spatial discretization

Two sets of angular and spatial discretization were designed. A fine one for the direct method and a coarser one for the perturbation method.

- First method: Difference of two k_{eff} calculations:
  To reach a specific worth convergence precision of 0.01 $10^{-5}/g$ the following numerical conditions were chosen. Typically, CALIBAN mesh size was 7200 cells without sample and 8216 cells with coated sample 1 (the little one). Considering the angular discretization, a S_{20} order was used. The angular scattering used a P_4 spherical harmonics decomposition.
- Second method: First order perturbation:
  The discretization conditions are less constraining with this method. The reactor mesh size was 525 cells without sample and 700 cells with coated sample 1. A S_{16} order angular discretization was used. The scattering kernel was treated using a P_4 spherical harmonics decomposition.

4.1.4 Multigroup discretization

A 48 energy group neutron cross-section file was produced using cross section processing system developed at CEA-Bruyères-le-Châtel using original ENDF/B-VI continuous energy library. The 48-group energy structure is obtained by even subdivision of a 16-group discretization. The 16-group energy structure is given in table 4. The spectral weighting function is a ^{235}U fission Watt spectrum.

4.1.5 Computation time optimization

Computation was accelerated using initial convergence on a coarse spatial mesh. A speed-up factor of about two was obtained by this way. Considering parallelization with energy group decomposition a speed-up factor greater than 10 was obtained with 24 processors when applied to the difference method which uses finest discretization.

4.2 Monte Carlo Calculations Using MCNP4C

We have used MCNP4C [7] code because it is a versatile and powerful tool concerning neutron transport. For each configuration, ENDF BVI continuous energy cross sections were used (.60c). The criticality calculations and track length estimates of the reaction rates used 5000 active generations (kcode and f4-fm4 options) with 5000 source particles per generation. We have skipped 50 generations for each calculation in order to have an accurate convergence of the fission source. Such computations were performed on the TERA machine (CEA-DAM Ile de France Simulation Project) wich is the most powerful computer in Europe.

The "track length estimation method" as explained in the MCNP4C manual may be extended in the following way to calculate the reactivity worth $\Delta\rho$ as defined in section 2.4 .

K_{sample} is the K_{eff} of the "reactor + Np sample".

$K_{coating}$ is the K_{eff} of the CALIBAN reactor with the coating.

P is a point in the phase space.

In the following relations, the K_{eff} are obtained after normalization with the source.

$$K_{sample} \equiv \int_{reactor} \nu\Sigma_f \Phi dP + \int_{N_p sample} (\nu\Sigma_f - \Sigma_a)\Phi dP$$

$$K_{coating} \equiv \int_{reactor} \nu\Sigma_f \Phi dP - \int_{coating} \Sigma_a \Phi dP$$

$$\Delta\rho = \frac{\frac{K_{sample}-1}{K_{sample}} - \frac{K_{coating}-1}{K_{coating}}}{M_{Np}}$$

We have verified that the discrepancy between the "classical" estimation of K_{eff} and the estimation given above is within the Monte carlo uncertainties, that is 0.00010 (relative uncertainty).

The scattering inside the iron coating or the sample is not taken into account because the outgoing energy of the neutrons is weakly modified (heavy media) and so we have made the assumption that such reactions were "fictitious".

Perturbation calculations using first and second order terms give very close results with the approach given above.

4.3 Results

We give in the table 5 the reactivity worths with ENDF BVI cross sections using MCNP4C and PANDA codes.
The masses correspond to the computer model, we recall in the table the experimental results, the worths are given in 10^{-5} per gram units. The relative uncertainties with MCNP4C and PANDA (direct method) are lower than 1 %.

There is a very good agreement between PANDA ENDF/B-VI calculations and experimental results. It can also be noticed that the specific worth increases with the sample mass because of a slight non-linearity of the perturbation.

There are discrepancies between MCNP4C and experimental results, that is close to 10 % (relative discrepancy). We may think that the approaches we have used ("Reaction rates" and Perturbation) were perhaps not designed for such configurations that is rigourous stepwise localised variations in material composition.
We can see that the overestimation decreases with the increasing of the sample mass.

TAB. 5 – **Neptunium-237 samples experimental and calculated specific worth.**

Mass (g)	w_{exp}	w_{PANDA}	w_{MCNP4C}
16.28	1.59	1.58	1.77
32.56	1.63	1.61	1.79
48.85	1.65	1.63	1.80

5 Conclusion

Numerical simulations of CALIBAN reactivity perturbation experiments using Np-237 samples were performed. A very good agreement was found between PANDA S_N code using ENDF/B-VI multigroup nuclear data and experimental results. The best agreement was found with the direct eigenvalue difference method which requires a very fine angular and spatial meshing together with a very restrictive convergence parameter. The first order perturbation method gives satisfactory results for small samples with coarser meshing conditions. Higher order perturbation should be used to account for the non-linearity of the reactivity with sample mass.

The Monte Carlo approach seems to be irrelevant because of the features of the problem.

We have also verified that the flux spectra were the same in the reactor and in the sample so the reactor imposes its spectrum to the Neptunium little sample. So we think that the experiments conducted by R Sanchez with a higher Neptunium mass are very interesting.

Calculations using JEF2 data have given results far from the experiment with every code. Those using JEF3 cross sections gave results close to results obtained with ENDF BVI data.

Since the experimental and PANDA (with ENDF BVI data) results were in very good agreement, we have used PANDA code to estimate the critical mass of ^{237}Np: 63.5 kg (ENDF BVI multigroup data used for the simulation of the experiment, density = 20.45 g/cm^3).

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Références

[1] R. G. Sanchez, "Critical Mass of ^{237}Np", *Proceedings of the Fifth International Conference on Nuclear Criticality Safety*, Albuquerque, NM, September 17-21 1995, pp. 182-189 (1995).

[2] J. Anno, G. Sert, "French Participation at ANS/ANSI 8/15 Working Group Updating Criticality Data on ^{237}Np Criticality and Transportation Proposals.", *ICNC'99*, Versailles, France, September 20-24 pp. 447-455 (1999).

[3] R. G. Sanchez, "Neptunium-237 and Highly Enriched Uranium Replacement Measurements Performed Using Flattop", NEA/NSC/DOC(95)03/VII Volume VII SPEC-MET-FAST-003.

[4] G. Assaillit, P. Zyromski, "Fast Burst Reactor CALIBAN", *Conference RADECS 99 (Radiation Effects on Components and Systems)*, Fontevraud, France, September 19-27 (1999).

[5] J. J. Duderstadt, W. R. Martin, *Transport Theory*, Wiley-Interscience Publication (1979).

[6] Ph. Humbert *Stochastic Neutronics with PANDA Deterministic Code*, M & C Meeting - Gatlinburg - April 2003.

[7] J. F. Briesmeister *MCNP4C Reference Manual*, LANL - CCC-700, April 2000.